

j.G. Tobin

February 12, 2010

#### Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

#### Recommendations

- 1. The present operating mode of the APS with regards to radioactive samples is a "distributed model," with access to many, if not all, beam-lines on the floor of the APS. The key is the utilization of containment, which is often triple containment, of small samples with limited radioactivity. The interaction of the APS Operations people, the APS Safety people, Lynne Soderholm and her ANL Chemistry people and the outside users has been spectacularly successful. It is essential that this mode of operation not be adversely affected by any new proposal or activity regarding radioactive samples at the APS.
- 2. On the other hand, there is the opinion amongst many uses that there is a need for an extended capability, for more highly radioactive or exposed actinide samples, that cannot be met within the distributed mode. Here, it is suggested that there is a need for a dedicated actinides building, adjacent to the APS but separated from it. This new building would incorporate several key components. Here is a brief synopsis: more detail will be given below.
  - a. Two beam-lines from the APS would run into the new building, with provision for sufficient isolation of the two buildings. Each of the two separate beam-lines would be supplied with photons by an undulator, on the APS ring. One would be a high-energy line, the other soft x-ray.
  - b. An extended capability for handling "bigger, hotter" and less contained radioactive samples would be included in this building, both on the beamlines themselves and in support laboratories. It would also include the capability for handling the influx of "bigger, hotter" samples from external facilities such as nuclear reactors.
  - c. The radioactivity levels within the new building would remain below Cat 3 levels, within the manifold of "radiation facilities" limits but above what is presently allowed at the APS.

The APS should pursue the construction a new, dedicated Actinides/Nuclear Fuels Facility, adjacent to the APS and including properly isolated beam-lines from the APS, as part of the APS Renewal.

Below, more detail and justifications will be provided in support of these recommendations.

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

# **Technological Motivation**

The world in general and the USA in particular are facing an oncoming energy shortage. One key mechanism to provide carbon-free energy is nuclear fission. At this point, 20% of the US electrical power grid is supplied by nuclear energy. [1] (Interestingly, it is 50% in Illinois. [2]) European nations such as Sweden (50% nuclear electricity [1]) and France (80% nuclear electricity [1]) are pushing ahead with permanent radioactive waste storage and processing. [3] If nothing else, the USA needs to provide the scientific foundation for improving its nuclear-power generation facilities. One key issue and how the APS could affect it are discussed below. (This discussion of this issue is not meant to be a comprehension argument in support of a facility but merely an example of the sort of science that could be pursued. An exhaustive collection of arguments would take more time and effort.)

The modification of various zones inside a nuclear fuel is an important issue. This includes microscopic re-crystallization, stress, fission gas production, He bubble formation and the intermixing, depletion and enrichment of various chemical, daughter and other isotopic species. For example, past studies [4,5] of the ternary nuclear fuel UPuZr have demonstrated constituent redistribution when irradiated or with thermal treatment

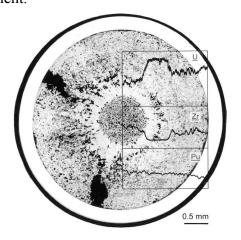


Figure 1
Postirradiation optical metallography and measured constituent redistributions in T179 fuel at 1.9 at.% burnup. Taken from Kim et al. Note the U depletion and Zr enrichment in the center zone and U enrichment and Zr depletion in the intermediate zone.

Energy Dispersive Spectroscopy (EDS) within an SEM was used to determine the concentration variations.

The concentration variations shown above are of significant concern. Driven in part by the thermal gradient within the nuclear fuel, these variations can affect reactor performance and fuel burn-up levels. Similar gradients were observed in samples that were not irradiated but underwent thermal gradient treatments. [5] From measurement such as these, kinetic parameters such as effective inter-diffusion coefficients were derived. The amount of such experimental data is very limited. Interaction of the fuel constituents with cladding and coolant are also important.

At present, INL scientists pursue a number of measurements on-site at INL and off-site to address issues such as this. [6] Here, we will propose two key examples of how a new facility at the APS could impact this technological issue.

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

# I Non-destructive cross-sectioning

In their talks at the workshop, Prof. Poulsen [7] and Dr. Ice [8] described some of the recent advances in imaging with hard x-rays. At this point in time, it seems that non-destructive cross-sectioning is a reality. An example from Prof. Poulsen is shown below.



Figure 2
This is a Diffraction Contrast Tomography (DCT) image, representative of two closely related techniques: 3DXRD and DCT. Specifically this is a rendition of the 3D grain structure in a cylindrical beta-Ti specimen containing 1008 grains, as obtained by the Diffraction Contrast Tomography. The photon energy was 40 KeV. From Ref 9 and courtesy of Prof. Henning Poulsen. Substantial improvements in spatial resolution may be possible, going into the nanoscale regime.

For example, one could address the issue of He bubble formation with a nanoscopic analysis of the elemental composition, stress, and ordering in an intact sample!

Using the Focussed Ion Beam (FIB) capabilities being developed at INL, it is conceivable that fairly small samples could be removed from the irradiated fuel pellets and sent to the APS/ANL for analysis. Size minimization would help to lower the total activity of the FIB samples, taken from the potentially highly radioactive, irradiated fuel pellet. Within this FIB sample, quantites such as the elemental composition, stress, and microcrystalline ordering could be measured with nanoscale accuracy.

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

# II Improved Simulation of Nuclear Materials via Experimental Benchmarking

In a break with past paradigms, the US Department of Energy has proposed a novel approach to the development of advanced nuclear fuels: predictive numerical simulation. [11] The advent of massive parallel computing and other improvements in computation capabilities has opened the door to the possibility of simulating much of the work that would have necessarily been determined empirically in the past. Nevertheless, these simulations and projections require the input of fundamental physical parameters that are experimentally generated or at least benchmarked. In particular, there is a dearth of fundamental thermodynamic information. To remedy this, we propose a radical departure from past practices of calorimetry. Using the techniques first proposed by Martensson and Johansson [12] and then validated by Steiner et al [13], we will use spectroscopically determined core level shifts to benchmark the computationally generated heats of solution. These measurements will be compared directly to the predictions of heats of solution from ab initio and CALPHAD calculations being conducted in complementary projects.

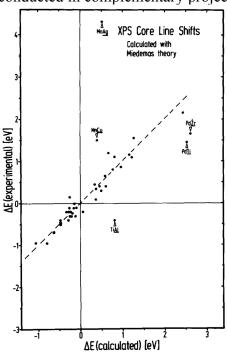


Figure 3

Plot of experimental binding energy shifts (y axis) versus the predicted values derived from heats of solution (x axis). Note the high degree of correlation, with only a few outriders away from the y = x line.  $YAXIS = \Delta E_{exp} = E_{alloy}(Z) - E_{pure}(Z)$  These are elementally specific experimental core binding energies. A is the dopant or solute, with atomic number Z. B is the host material or solvent.  $XAXIS = \Delta E_{calc} = E(Z,B) + E(Z+1,Z) - E(Z+1,B)$  These are heats of solution! Taken from Steiner et al. [13]

Owing to resolution (100meV bandpass) and intensity arguments (1% alloy compositions), only soft x-ray (hv = 500 - 1000 eV) photoelectron spectroscopy will work. Furthermore, this requires "exposed samples," where triple containment is impossible. In fact, the vacuum vessel becomes one level of containment and the others are provided a glove-box built around the vacuum vessel and the room itself, similar to the functional approach at the Institute for TransUranics (ITU), in Karlsruhe, Germany. [14] Further details of Spectroscopic Calorimetry can be found elsewhere. [15]

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

# **Conceptual Design**

- 1. Two slightly canted undulators in one straight section. A separation of a degree between the undulators would provide a lateral separation of 2.6 meters at 150 meters out.
- 2. Experiments on the two beamlines will be laid out in tandem fashion, one after the other.
  - a. High energy line
    - i. Non-destructive cross-sectioning/imaging (described above)
    - ii. **Structure and Coordination environment/ oxidation states as a function of temperature.** This would be the multi-technique approach for higher activity samples, e.g. nuclear fuels and cladding. Techniques would include x-ray diffraction, atomic pair distribution functions, XANES, and EXAFS, at low and high temperatures.
    - iii. **Micro-diffraction**. Here, work would be done on phase identification, inter-granular stress, sub-grain structure and radiation damage.
    - iv. **High Energy Photoelectron Spectroscopy (hv = 6 KeV).** Bulk electronic structure and density of states.
  - b. Soft X-ray line
    - i. Spectroscopic Calorimetry. (described above)
    - ii. **Fano Spectroscopy.** Electronic structure of the 5f's, including electron correlation, using solid and evaporated actinide samples.
    - iii. **Magnetism in Actinides.** X-ray Magnetic Circular Dichroism in X-ray Absorption Spectroscopy (XMCD-XAS) and related soft x-ray techniques.
- 3. **Materials Handling of "bigger, hotter" samples**: Here the limits will be driven by the beam, not the sample, but the sample will remain below Category 3 limits.
  - a. High activity samples. These could be macroscopic, e.g. a disc cut from a fuel rod or pellet, with a 10mm diameter and a 1 mm thickness, or larger cladding and component samples.
  - b. Micro-manipulation/nano-positioning- in situ, on line.
  - c. Off line sample handling, storage, shipping/receiving of irradiated samples.
  - d. A Rad Lab for hotter samples, with a standard suite of analytical techniques and support measurements.
  - e. Liquid sample and gas flow reaction handling on the beam-line.
  - f. Rad sample heating, molten Pu desired –on line
  - g. An on-site staff of specialized and dedicated support personnel.
  - h. Irradiation capability-ion beam bombardment on line.
  - i. In situ sample conditioning. This would include determination of the carbon and oxygen potentials of carbides, oxides and oxycarbides of Pu and U.

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

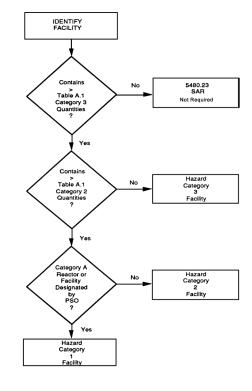
#### Discussion of radiation levels

It is our opinion that the new building should be a radiological facility but not Category, 1, 2, or 3. Category 3 has the lowest threshold. First we will look at the guidance and the decision making flow chart.

#### Category 3 Thresholds

DOE-STD-1027-92

Figure 3.1 HAZARD CLASSIFICATION DECISION PROCESS (Section 3



# Guidance from DOE-STD-1027-92, page 2 [16] 1.0 SAR NUCLEAR FACILITY GUIDANCE FOR DOE ORDER 5480.23

Order 5480.23 defines the "level of concern" within the framework of Hazard Categorization, which requires the preparation of a SAR for DOE nuclear facilities. Section 3 and Attachment 1 of this Standard provide consistent guidance on facility categorization. All facilities classified as at least Category 3 in accordance with this guidance are required to comply with DOE Order 5480.23. Additional guidance regarding some environmental restoration activities is provided in an Interpretation Memo dated June 9, 1997, Black to Psaras. Facilities that do not meet or exceed Category 3 threshold criteria but still possess some amount of radioactive material may be considered Radiological Facilities.

Figure 4, To the Left: Decision Process Diagram from Page 8 of Ref 16.

# **Determination of the Category 3 Thresholds**

8

DOE-STD-1027-92 provides guidance concerning the translation of Category 3 guidelines into amounts of specific elemental materials and isotopes. Further guidance can be found in Ref 17. In Reference 17, there are example calculations of the amounts of specific materials that correspond to the Category 3 threshold. On the following page, two specific calculations are shown, for Fuel Grade Pu and Weapons Grade Pu. The key is that minority constituents can have a dramatic effect because of their heightened activity. To quote Ref 17: "Applying this technique (Eq. 3) to plutonium, the value for the Category 3 mass limit is found to be 2.65 g for fuel grade material (18 wt. % 240Pu) and 5.67 g for weapons grade (see Tables 1. & 2.). Note that this last value differs significantly from the value of 8.4 gm if the isotopic value, as listed in Table A.1, for 239Pu is used. The reason is that the impact of the 240Pu and 241Am activity is still significant." This issue is very important for irradiated samples.

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

Table 1. Determination of Mix Category 3 Mass Threshold Value-Pu Fuel Grade.

Isotope	fi	SAi	Ti (Ci)	fi x SAi	(fi xi) \$ATi
	(wt. % /100)	(Ci/gm)		(Ci/gm)	(1/g)
238 <sub>Pu</sub>	0.001	17.0	0.62	0.017	0.027
239 <sub>2</sub> u	0.78	0.062	0.52	0.048	0.092
249 <sub>u</sub>	0.18	0.23	0.52	0.041	0.079
241 <sub>Pu</sub>	0.016	103.0	32.0	1.648	0.052
242 <sub>Pu</sub>	0.0049	0.004	0.62	0.000	0.0
24 <u>1</u> Am	0.019	3.47	0.52	0.066	0.127
Σ					0.377
					1

$$Q = \frac{1}{0.377} = 2.65g$$

Table 2. Determination of Mix Category 3 Mass Threshold Value-Pu Weapons Grade.

Isotope	fi	SAi	Ti (Ci)	f <sub>i xi</sub> SA	(fixi) SATi
	(Wt. %/100)	(Ci/gm)		(Ci/gm)	(1/g)
238 <sub>Pu</sub>	0.00306	17.0	0.62	0.0061	0.010
23 <b>%</b> u	0.933	0.062	0.52	0.0578	0.111
240-u	0.0599	0.23	0.52	0.0138	0.026
24 þu	0.00282	103.0	32.0	0.2905	0.009
242 <sub>pu</sub>	0.0004	0.004	0.62	0.000	0.0
24 Am	0.00294	3.47	0.52	0.0102	0.010
Σ					0.176

$$Q = \frac{1}{0.176} = 5.67g$$

# Q = Quantity of material used as threshold (grams)

DOE-STD-1027-92 ATTACHMENT 1

Isotope	Category 2 Curies	Threshold Grams	Category 3 Curies	Threshold Grams
U-233	2.2E+02***	2.3E+04***	4.2E+00	4.4E+02
U-234	2.2E+02	3.5E+04	4.2E+00	6.7E+02
U-235	2.4E+02***	1.1E+08***	4.2E+00	1.9E+06
U-238	2.4E+02	7.1E+08	4.2E+00	1.3E+07
Np-237	5.8E+01	8.3E+04	4.2E-01	6.0E+02
Np-238	9.1E+05	3.5E+00	1.3E+03	5.0E-03
Pu-238	6.2E+01	3.6E+00	6.2E-01	3.6E-02
Pu-239	5.6E+01***	9.0E+02***	5.2E-01	8.4E+00
Pu-241	2.9E+03	2.8E+01	3.2E+01	3.1E-01
Am-241	5.5E+01	1.6E+01	5.2E-01	1.5E-01
Am-242m	5.6E+01	5.8E+00	5.2E-01	5.3E-02
Am-243	5.5E+01	2.8E+02	5.2E-01	2.6E+00
Cm-242	1.7E+03	5.1E-01	3.2E+01	9.7E-03
Cm-245	5.3E+01	3.1E+02	5.2E-01	3.0E+00
Cf-252	2.2E+02	4.1E-01	3.2E+00	5.9E-03

Figure 5(above) Some representative Category 3 limits from Ref 17.

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

Below is shown a brief summary of the limits at some synchrotron radiation facilities. It is clear that these limits are substantially below the Category 3 threshold.

Provided by Gerry Lander and presented at the APS Rad Workshop in Jan 2010

# **Limits at synchrotrons**

It depends on countries, and it depends how the rules are interpreted by the radiation safety officers at the source.

The numbers below are based on  $^{239}$ Pu equivalent specific activity; they are less than at the neutron centers by at least an order of magnitude and in some cases (ESRF & ILL, Grenoble) by a factor of  $10^6$ . 1 g  $^{239}$ Pu = 2.4 GBq.

SSRL(US): 10 GBq Certainly the largest anywhere at the moment!

APS (US): 0.6 GBq (250 mg <sup>239</sup>Pu): NSLS would be same

ALS (US): no transuranium allowed (soft x-rays)

ESRF (F): 0.2 GBq at the Rossendorf beamline; 0.0004 GBq at other beamlines

**ANKA** (D):  $\sim 0.004$  GBq but no <sup>239</sup>Pu or <sup>235</sup>U (no fission isotopes)

DIAMOND (UK) & SLS (Sw): essentially zero! Panic over depleted U

**SOLEIL** (**F**): MARS beamline (2011) will allow 18.5 GBq

Note: The operational limit at the ALS appears to be micrograms of Pu. Only microscopy experiments are allowed.

Figure 6 (above) Some limits for rad materials. [18]

Nuclide	Specific	Derived Air	Maximum	Maximum Allowed
Activity	Solid	Concentration	Allowed	Solid Sample
	Sample	(μCi/cc)	Solid Sample	Weight
	Activity *		Activity	(g)
	(Ci/g)		(μCi)	
Th-229	2.12E-01	4.0E-13	3	1.415E-05
Th-230	2.05E-02	3.0E-12	24	1.17E-03
Th-232	1.09E-07	5.0E-13	4	3.67E+01
U-235	2.15E-06	2.0E-11	163	7.58E+01
U-238	3.35E-07	2.0E-11	163	4.87E+02
Nat-U	6.85E-07	2.0E-11	163	2.38E+02
Dep-U	3.35E-07	2.0E-11	163	4.87E+02
Np-237	6.99E-04	2.0E-12	16	2.29E-02
Pu-238	17.0	3.0E-12	24	1.41E-06
Pu-239	6.19E-02	3.0E-12	24	3.88E-04
Pu-240	2.27E-01	3.0E-12	24	1.06E-04
Pu-242	3.91E-03	3.0E-12	24	6.14E-03
Am-241	3.42	3.0E-12	24	7.02E-06
Am-243	0.198	3.0E-12	24	1.21E-04
Cm-248	4.23E-03	7.0E-13	6	1.42E-03
Cf-248	1.58E03	3.0E-11	244	1.5E-07
Cf-252	5.35E02	8.0E-12	65	1.22E-07
Bk-249	1.63E03	7.0E-10	5691	3.49E-06
Es- 253	2.51E04	6.0E-10	4878	1.9E-07
Sr-90	13.7	8.0E-09	6.5E04	4.74E-03
Tc-99	1.69E-02	3.0E-07	2.4E06	1.42E+02
Tc-99m	5.24E06	6.0E-05	8.13E07	1.55E-05

Table 1: Maximum Allowed Solid Sample Activity of Selected Radionuclides

Figure 7 (above) Published limits for rad materials at the APS. [19]

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

However, we need to take a closer look at the limits for rad work at the APS. For the sake of argument, this discussion will be cast in terms of the amount of 239Pu allowed.

	Mass <sup>239</sup> Pu(mg)	% of Th. Lim.	% of Cat 3 Lim.	Improvement
LIMIT			(5,670mg)	Factor
	A	B = A/250mg	C = A/5,670mg	D = 1/C
Theoretical	250	100%	4%	20
APSWebsite[19]	0.4	0.2%	0.007%	14,000
Recent Exp[20]	5	2%	0.09%	1,100

#### Table 1

Table 1 Caption The theoretical limit is taken from Figure 6. The website value is taken from the documentation provided on the APS website. [19] This guidance may be slightly dated but it is the point at which negotiations begin. The Recent Experiment comes from an x-ray diffraction experiment. [20] Here, the more conservative estimate of 5.67g of weapons grade 239Pu is used instead of the 8.4 g for pure 239Pu. However, both values would give similar results.

Compare to the APS guidelines on the APS web-site, as shown in Figure 7, the improvement factor in going to a Cat 3 limit would be 14,000. This is obviously too high. Similarly, the theoretical value gives an improvement factor of 20. While even an improvement factor of 20 would be worth pursuing, this estimate is too conservative, because there is no indication that the theoretical limit has been approached. Instead, the largest amount of 239Pu allowed on the floor to date has been on the scale of 5 mg. [20] The improvement factor here would be on the scale of 1000.

These arguments do not broach an equally important subject: irradiated samples and heated samples. Both the radiologically hot and thermally hot Rad samples are hampered by further restrictions that could be alleviated by a new, dedicated building.

#### Some Case Studies and other Input

#### An example of a previous experiment at the APS upon an irradiated sample.

The technical details of the experiment can be found in Ref 21-25. The fuel was ATM-106, discharged in 1980 (so about 26 year out-of reactor), with rather high burn-up, about 42 MWd/kgU. It was calculated that there was about 1 mg of fuel, containing 6 MBq. The dose rate was about 1 mR/h at 30 cm. There were minor issues with biological shielding, to maintain public spaces below the allowable radiation field limits when the specimen was in storage outside the hutch. Once inside the hutch, security was a concern owing to the nature of the sample and the fact that the entire hutch was a radiologically controlled area, to be accessed only by approved DOE-certified radiation workers. [26] Thus a 1mg activated sample was allowed. For many experiments, this size may be a limitation.

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

# **Examples of higher activity samples**

For a 1 mm thick disc (of 10 mm diameter) of UO2 fuel the activity will vary between about 5 and 15 GBq depending on the burn-up. (5 GBq at around 10 MWd/kgHM and 15 GBq at 70 Mwd/kgHM.) It is assumed that a 1 mm thick disc contains 0.07 g of fuel and the activity of the surrounding cladding has been neglected. (It will be small in comparison with the fuel). The cooling time (interval between discharge from the reactor and date when the activity was determined) is 7 to 9 months. As to the dose rate, for 0.1 g of UO2 fuel with a burn-up of about 50 MWd/kgHM the contact dose rate will be about 7 R/h after cooling for 1 or 2 years. [27]

#### **Example of LANL Experiments in support of INL Nuclear Fuel Development.**

There is a need for 100-200 micron thick Pu in 1-5 mm diameter (thin cylinder type samples) for diffraction both in transmission and diffraction, the weight would be in the 40-250 milligram range. It should be possible to get transmission through 100 microns on a high energy beamline (like the 1ID which is in the 80-100keV range). It is probable that one could get micron or microns scale strain, phase and orientation info with microbeam techniques at high energy, and at lower energy submicron scale information on much much smaller samples (15-30microns thick). Just for sample handling issues it would help to have a mm or more diameter even on the really thin samples. It would be good to have the ability to do similar scanning on oxide and metallic fuel pellets, the density of which would probably be about half of alpha Pu. A rough estimate is that guess that the x-rays can get through 200 microns. In this case, it would be good to have the ability to do full pellets at 5-10mm diameter by whatever thickness can be transmitted through. Again, twice the thickness probably, but half the density gives the same ball park measurement for the same type of grain mapping, micron scale sub grain info. The idea would be to get strain and microstructural evolution across the length scale of the inhomogeneities. In both the as sintered pellets and the PI this can go across the entire sample pellet. It would be great, as mentioned in the meeting, to be able to look at a slice of pellet with the cladding on it, so we could also probe that interface change at the micron level. [28]

While the 250 mg mass is within the theoretical APS limits, it is well outside of the established operation maximum to date of 5 mg.

Based upon discussions with Stu Malloy and other cladding/irradiated steel sample people, similar experiments to fuels and Pu would need to happen across a 3mm length scale, which would be the thickness of some of the ducts around reactor bits. Again, 3mm thick samples x 1mm would be great for transmission experiments. These samples would have a 239 equivalent as they are irradiated to sometimes 100's of DPA and activate to isotopes that can be equated to 239. Some of these small samples (3mmx1mmx1mm) that we deal with can have dose measurements at contact as high as 500millirem/hr. Many of the smaller samples are much more reasonable in the 2-20 millirem/hour but some of the

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

samples fresh out of reactors may be quite high. These would all fall under the heading of PIE post-irradiation experiments. [28]

# **Input from Gerry Lander [29]**

Since the APS workshop last week, I think there is an important development. In two email exchanges, the first below, the second as the last attachment, you will see that experiments on small pieces of irradiated fuel have been done at APS, and that experiments on bigger pieces that could be of major interest to the nuclear program could be done if the limits were higher. The experiments reported by Jeff Fortner and his collaborators are clearly of great interest. These have been done under the APS limit of 0.6 GBq of inventory. They, of course, will continue, although the closing of the hot cells at CHM at ANL is a severe drawback. Presumably, some preparation could be done at INL. However, if that limit was extended to 18.5 GBq (which is what the French presently propose at the MARS beamline at Soleil) then larger pieces could be examined. Experiments like imaging, phase identification, and elemental analysis (as already performed by Fortner et al) would become available on a wider scale. Such an extension would, I believe, require a separate building and beamline.

I do not know the lower limits of cat III, but I doubt they are this low - this represents < 100 mg of spent fuel. However, that needs to be checked. To get anything done would need the full cooperation of NE, but I do believe, especially from what I hear from the people at Karlsruhe who work with such fuel, that the information would be of great value, not only for present spent fuels, but for proposed fuels for the future. The French, as far as I could gather, plan to have capability of powder diffraction and EXAFS, but I think APS should go further than this and include imaging. I send you all this as I had the impression that a strong case for a special beam was not developed at the Workshop, but part of that was the understandable reluctance to consider a cat III facility, and the lack of knowledge of how much activity was involved in spent fuel samples. I think we have resolved that problem. The issue of whether the facility would be useful to fusion research is still open.

Hoping this and the attachments are useful. Best regards Gerry

#### Conclusions

The likelihood that operations at the APS will approach the 250mg limit, much less the Cat 3 limit, seem small. There are also indications of the need facility, including beamlines, for samples that are "bigger and hotter." Thus, the recommendation is to build a new, dedicated actinides and nuclear fuels building at the APS with undulator beamlines and enhanced sample handling capabilities.

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

#### References

- 1. Nuclear Energy Institute, Nuclear shares of electricity generation, <a href="http://www.world-nuclear.org/info/nshare.html">http://www.world-nuclear.org/info/nshare.html</a>
- 2. US Energy Information Administration, State Nuclear Industry Illinois, <a href="http://www.eia.doe.gov/cneaf/nuclear/page/at-a-glance/states/statesillinois">http://www.eia.doe.gov/cneaf/nuclear/page/at-a-glance/states/statesillinois</a>
- 3. SKB-Sweden website, <a href="http://www.skb.se/default">http://www.skb.se/default</a>
- 4. P. Steiner, S. Huefner, M. Martensson and B. Johansson, Solid State Communications **37**, 73 (1981).
- 5. Y.S. Kim, G.L. Hofman, S.L. Hayes and Y.H. Sohn, J. Nucl. Matl. **327**, 27 (2004).
- 6. Y.H. Sohn, M.A. Dayananda, G.L. Hofman, R.V. Strain and S.L. Hayes, J. Nucl. Matl. **279**, 317 (2000).
- 7. Jon Carmack (INL), "Fuel Cycle R & D Advanced Fuel Development," APS Rad Workshop, January 2010, http://www.aps.anl.gov/News/Conferences/.
- 8. Henning Poulsen (DTU-Risø), "4D tools for studying microstructure dynamics," APS Rad Wksp, Jan 2010, http://www.aps.anl.gov/News/Conferences/.
- 9. Gene Ice (ORNL), "Microbeam, timing, and wavelength dispersive studies of nuclear materials using 3<sup>rd</sup> and 4<sup>th</sup> generation synchrotron sources," APS Rad Workshop, January 2010, http://www.aps.anl.gov/News/Conferences/.
- 10. W. W. Ludwig, P. Reischig, A. King, M. Herbig, E. M. Lauridsen, G. Johnson, T. J. Marrow, and J. Y. Buffière Rev. Sci. Instruments **80**, 33905 (2009).
- 11. D.A. Petti, S, Hayes, F. Garner, S. Maloy, B. Wirth, and S. Zinkle, "Department of Energy Review Committee Report on the Review of Irradiation Testing facilities for the Global Nuclear Energy Partnership," August 2006.
- 12. N. Martensson and B. Johansson, Solid State Comm. **32**, 791 (1979)
- 13. P. Steiner, S. Huefner, M. Martensson and B. Johansson, Solid State Communications **37**, 73 (1981).
- 14. L. Havela, T. Gouder, F Wastin and J. Rebizant, Phys. Rev. B **65**, 235118 (2003).
- 15. Spectroscopic Calorimetry, J.G. Tobin, <a href="http://gurka.fysik.uu.se/esp/">http://gurka.fysik.uu.se/esp/</a>, <a href="http://gurka.fysik.uu.se/workshop/workshop.html">http://gurka.fysik.uu.se/workshop/workshop.html</a>.
- 16. DOE-STD-1027-92, December 1992, CHANGE NOTICE NO.1 September 1997, DOE STANDARD, "HAZARD CATEGORIZATION AND ACCIDENT ANALYSIS TECHNIQUES FOR COMPLIANCE WITH DOE ORDER 5480.23, NUCLEAR SAFETY ANALYSIS REPORTS," U.S. Department of Energy, AREA SAFT, Washington, D.C. 20585.
- 17. "Estimating Inventory Thresholds for Nuclear Facilities Using DOE STD-1027-92 Attachment 1 Table A.1 'Thresholds for Radionuclides'," D. Price, J.S. Hildum and A.C. Williams, Safety Analysis Transitions Workshop, Oakland, CA, June 9-13, 1997, UCRL-JC-127245, LLNL Library Catalog, http://library-r.llnl.gov/uhtbin/cgisirsi/mw0o9EOMyb/MAIN/9171
- 18. Gerry Lander (ITU, retired), "Research on transuranium systems: present capabilities with large central facilities," APS Rad Workshop, January 2010,

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

- http://www.aps.anl.gov/News/Conferences/.
- 19. "Policy for Conducting Radioactive Sample Experiments in APS Experiment Enclosures, March 15, 2004 (revised May 17, 2005) available at <a href="https://www.aps.anl.gov/Safety\_and.../Experiment\_Hazard.../RadSamplepolicy.pdf">www.aps.anl.gov/Safety\_and.../Experiment\_Hazard.../RadSamplepolicy.pdf</a>.
- 20. Private Communication, Mark Wall, LLNL, February 2010.
- 21. A. J. Kropf, J. A. Fortner, R. J. Finch, J. C. Cunnane and C. Karanfil, Physica Scripta. Vol. T115, 998–1000, 2005.
- 22. JEFFREY A. FORTNER, ROBERT J. FINCH, A. JEREMY KROPF, and JAMES C. CUNNANE, NUCLEAR TECHNOLOGY 148, 174 (2004).
- Jeffrey A. Fortner, A. Jeremy Kropf, James L. Jerden, and James C. Cunnane, Scientific Basis for Nuclear Waste Management XXX, edited by D.S. Dunn, C. Poinssot, B. Begg (Mater. Res. Soc. Symp. Proc. 985, NN01-03 (2007).
- 24. J.A. Fortner, A.J. Kropf, R.J. Finch, A.J. Bakel, M.C. Hash, D.B. Chamberlain, Journal of Nuclear Materials 304, 56–62 (2002).
- 25. Jeffrey A. Fortner, A. Jeremy Kropf, Robert J. Finch, and James C. Cunnane, Scientific Basis for Nuclear Waste Management XXVIII, edited by John M. Hanchar, Simcha Stroes-Gascoyne, and Lauren Browning, Mater. Res. Soc. Symp. Proc. **824**, CC2.11. P. 107–112 (2004).
- 26. Private Communication, Jeffrey A Fortner, Ph.D., Argonne National Laboratory, Chemical Sciences and Engineering Division,m Feb 2010.
- 27. Private communication, Clive Walker, ITU, <u>Clive.WALKER@ec.europa.eu</u>, Feb 2010
- 28. Private communication, Tarik Saleh, LANL, February 2010.
- 29. Private Communication, Gerry Lander, ITU, February 2010.

Report of the Bulk Working Group
Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

# Appendix: Viewgraphs from the Bulk Working Group

Report of the Bulk Working Group
Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear
Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

Viewgraphs pre	nne Natl Lab VG # 2 of 3	
Bulk Working Group Techniques-corresponding to page 1	In situ sample conditioning	Total of 3 slides

Report of the Bulk Working Group
Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear
Energy Systems. 27-28 January 2010, Advanced Photon Source, Argonne Nat'l Lab, Chicago, IL, USA

ples		nm imples		mples	nes and					က
Bulk Working Group Materials handling of big, hot samples -limits driven by beamline, not sample,	but less than Category 3	High activity samples, macroscopic, e.g fuel rod/pellet disc with 10 mm diameter and 1 mm thickness and larger cladding and component samples	Micro-manipulation, nano-positioning –in situ	Off-line sample handling, storage, shipping/receiving of irradiated samples	Rad Lab for hotter samples, with standard suite of analytical techniques and support measurements	Liquid sample and gas flow reaction handling on the beamline	Rad sample heating; molten Pu desired	Specialized support personnel	Irradiation capability (Ion beam bombardment online)	Total of 3 slides
Mate-limit		High a	Sign -	\$ *	• Rad La		• Rad sa	* Special		